

Calibration of the Lawrence Livermore National Laboratory Passive-Active Neutron Drum Shuffler for Measurement of Highly Enriched Uranium in Mixed Oxide

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Calibration of the Lawrence Livermore National Laboratory Passive-Active Neutron Drum Shuffler for Measurement of Highly Enriched Uranium in Mixed Oxide

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Abstract

As a follow-on to the Lawrence Livermore National Laboratory (LLNL) effort to calibrate the LLNL passive-active neutron drum (PAN) shuffler for measurement of highly enriched uranium (HEU) oxide, a method has been developed to extend the use of the PAN shuffler to the measurement of HEU in mixed uranium-plutonium (U-Pu) oxide. This method uses the current LLNL HEU oxide calibration algorithms, appropriately corrected for the mixed U-Pu oxide assay time, and recently developed PuO_2 calibration algorithms to yield the mass of ^{235}U present via differences between the expected count rate for the PuO_2 and the measured count rate of the mixed U-Pu oxide. This paper describes the LLNL effort to use PAN shuffler measurements of units of certified reference material (CRM) 149 [Uranium (93% Enriched) Oxide - U_3O_8 Standard for Neutron Counting Measurements] and CRM 146 [Uranium Isotopic Standard for Gamma Spectrometry Measurements] and a selected set of LLNL PuO_2 -bearing containers in consort with Monte Carlo simulations of the PAN shuffler response to each to (1) establish and validate a correction to the HEU calibration algorithm for the mixed U-Pu oxide assay time, (2) develop a PuO_2 calibration algorithm that includes the effect of PuO_2 density (2.4 g/cm^3 to 4.8 g/cm^3) and container size (8.57 cm to 9.88 cm inside diameter and 9.60 cm to 13.29 cm inside height) on the PAN shuffler response, and (3) develop and validate the method for establishing the mass of ^{235}U present in an unknown of mixed U-Pu oxide.

Introduction

The logical follow-on to the Lawrence Livermore National Laboratory (LLNL) effort to calibrate the LLNL passive-active neutron drum (PAN) shuffler for the measurement of highly enriched uranium (HEU) oxide was the calibration of the PAN shuffler for the measurement of HEU in mixed uranium-plutonium (U-Pu) oxide. The method that was developed uses the current LLNL HEU oxide calibration algorithms, appropriately corrected for the mixed U-Pu oxide assay time, and recently developed PuO_2

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calibration algorithms to yield the mass of ^{235}U present via differences between the expected count rate for the PuO_2 and the measured count rate of the mixed U-Pu oxide.

Calibration Plan

The components of the calibration plan include (1) the development of PuO_2 working reference materials (WRMs), (2) the determination of an appropriate mixed U-Pu oxide assay time, (3) the effect of the mixed U-Pu oxide assay time on the HEU oxide calibration algorithms, (4) the replicate measurement of PuO_2 WRMs, (5) Monte Carlo simulations of the PAN shuffler response to the PuO_2 WRMs, (6) Monte Carlo simulations of the PAN shuffler response sensitivity to variations in PuO_2 density and container diameter, (7) the development of a set of PuO_2 calibration algorithms and their associated errors, and (8) the development of mixed U-Pu oxide calibration algorithms and their associated errors.

Development of PuO_2 Working Reference Materials

The need for PuO_2 calibration algorithms necessitated the development of PuO_2 WRMs from the well characterized LLNL inventory of PuO_2 -bearing containers. Items developed as WRMs were selected on the basis of their lack of contaminants as reported in the LLNL special nuclear material database and their packaging in either of two primary containers: Kaufman can (8.57 cm inside diameter and 9.60 cm inside height) and quart can (9.88 cm inside diameter and 13.29 cm inside height). A minimum of three replicate calorimeter and gamma isotopic counter measurements were used to establish the total Pu mass and isotopic composition (^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am , and ^{237}Np) of each PuO_2 WRM.

Determination of an Appropriate Mixed U-Pu Oxide Assay Time

The current LLNL HEU oxide calibration algorithms were developed from replicate PAN shuffler measurements of units of certified reference material (CRM) 149 [Uranium (93% Enriched) Oxide - U_3O_8 Standard for Neutron Counting Measurements] and CRM 146 [Uranium Isotopic Standard for Gamma Spectrometry Measurements] in consort with Monte Carlo simulations of the PAN shuffler response to each [1]. A standard U oxide assay time was used for each measurement, consisting of a nominal 270 s background count followed by 34 shuffles (cycles of irradiation and counting), each with a 20.8 s period (a 1.4 s forward transfer time, an 11.7 s ^{252}Cf source irradiation time, a 0.8 s reverse transfer time, and a 7 s count time).

Initial PAN shuffler measurements of the PuO_2 WRMs under the U oxide assay conditions resulted in delayed neutron count rates with signal to noise ratios not appreciably greater than one and large counting errors. As such, a parameter study was initiated using the basic equation for the PAN shuffler delayed neutron count rate [2] to determine a more appropriate standard assay time for PuO_2 -bearing materials. The only constraint on the study was a measurement consideration requirement that the total assay time not exceed one hour. The standard Pu oxide assay time that resulted consisted of a nominal 1080 s background count followed by 68 shuffles (cycles of irradiation and counting), each with a 29.2 s period (a 1.4 s forward transfer time, a 20.0 s ^{252}Cf source irradiation time, a 0.8 s reverse transfer time, and a 7 s count time). Subsequent measurements of the PuO_2 WRMs under the Pu oxide assay conditions resulted in delayed neutron count rates with improved signal to noise ratios and reduced counting errors. Furthermore, the average of three consecutive replicate measurements of a given item were found to reduce the overall error even more.

Suffice it to say that the Pu oxide assay time was adopted for the measurement of mixed U-Pu oxide items along with the practice of using the average of three consecutive replicate measurements of a given item.

Effect of the Mixed U-Pu Oxide Assay Time on the HEU Oxide Calibration Algorithms

Selection of a mixed U-Pu assay time that differed from the U assay time used in development of the HEU oxide calibration algorithms necessitated an evaluation of the effect of the mixed U-Pu assay time on the delayed neutron count rate. To this end, a minimum of five consecutive measurements were made of each container in the LLNL units of CRM 146 and CRM 149 under the mixed U-Pu assay conditions. An average delayed neutron count rate was computed for each and compared with the associated average delayed neutron count rate measured under the U assay conditions. Table 1 summarizes the comparison of the mixed U-Pu oxide assay time and U assay time measured delayed neutron count rate results for the LLNL units of CRM 146 and CRM 149.

Table 1. Comparison of the mixed U-Pu assay and U assay measured delayed neutron count rate results for the LLNL HEU oxide standards.

LLNL Nomenclature	U-Pu assay measured count rate (counts/s)	U assay measured count rate (counts/s)	Measured U-Pu assay to U assay ratio
CRM146-20	96.88 ± 0.91	86.49 ± 1.42	1.120 ± 1.89%
CRM146-52	187.364 ± 0.93	165.70 ± 2.28	1.131 ± 1.46%
CRM146-93	273.873 ± 1.22	240.91 ± 1.34	1.137 ± 0.71%
CRM149-05	618.350 ± 1.92	557.19 ± 6.19	1.113 ± 1.15%
CRM149-10	1118.54 ± 1.72	1000.88 ± 5.14	1.118 ± 0.54%
CRM149-15	1579.46 ± 2.82	1411.18 ± 5.21	1.119 ± 0.41%
CRM149-20	2033.06 ± 3.44	1824.29 ± 6.81	1.114 ± 0.41%
CRM149-30	2959.90 ± 4.23	2650.21 ± 8.54	1.117 ± 0.35%
CRM149-40	3865.20 ± 10.98	3459.95 ± 8.62	1.117 ± 0.38%
Average			1.121 ± 0.79%

From the above, it follows that the delayed neutron count rate predicted for the U mass in a mixed U-Pu oxide must be reduced by a factor of 1.121 before using the HEU oxide calibration algorithms to predict the measured U mass.

Measurement of the PuO₂ Working Reference Materials

Each LLNL developed PuO₂ WRM received a minimum of three consecutive replicate PAN shuffler measurements. Table 2 summarizes the pertinent mass and isotopic composition information for the LLNL developed PuO₂ WRMs at the PAN shuffler measurement (reference) date.

Monte Carlo Simulations of PAN Shuffler Response to PuO₂ Working Reference Materials

Monte Carlo simulations of the PAN shuffler response to the PuO₂ WRMs were performed with the MCNP code [3] using the technique and post-processor developed by Rinard [4]. Models of the PuO₂ WRMs used in the MCNP simulations were true to the packaging of each, including the primary containers, packing materials polyvinylchloride bag-out bag and polyethylene poultry bag), and secondary (over-pack) containers required to satisfy LLNL Plutonium Facility containment requirements. Because the PuO₂ density of each WRM was

Table 2. Pertinent mass and isotopic composition information for the LLNL PuO₂ WRMs.

LLNL Nomenclature	Reference date	PuO ₂ mass (g)	Pu mass (g)	²³⁹ Pu wt% (%)	²³⁹ Pu mass (g)	²⁴⁰ Pu mass (g)	²⁴¹ Pu mass (g)
PuOKC0040	10/19/2000	53.16	46.89	93.9109	44.03	2.79	0.06
PuOKC0080	10/23/2000	96.29	84.92	93.9086	79.75	5.05	0.09
PuOSQ0110	5/19/2000	128.24	113.10	94.6790	107.08	5.93	0.06
PuOKC0160	10/16/2000	187.21	165.12	93.9752	155.17	9.57	0.32
PuOKC0210	10/12/2000	250.89	221.28	93.9403	207.87	13.08	0.26
PuOKC0240	10/4/2000	291.20	256.83	93.8982	241.16	15.25	0.33
PuOSQ0340	6/2/2000	402.98	355.42	93.9421	333.89	20.97	0.45
PuOKC0420	10/18/2000	497.96	439.19	93.9436	412.59	25.89	0.57
PuOSQ0610	6/23/2000	725.87	640.20	94.9050	607.58	32.06	0.40
PuOSQ0800	5/26/2000	966.01	852.00	93.9262	800.25	49.74	1.69
PuOKC0810	10/6/2000	974.78	859.73	93.9439	807.67	50.47	1.31
PuOKC1010	10/5/2000	1212.11	1069.04	93.8674	1003.49	63.27	1.92
PuOSQ1090	5/26/2000	1313.20	1158.20	94.0696	1089.52	66.88	1.47
PuOKC1250	10/17/2000	1513.60	1335.03	93.9450	1254.20	78.23	2.13
PuOSQ1410	5/25/2000	1693.09	1493.26	93.9980	1403.64	87.73	1.40
PuOSQ1640	9/29/2000	1969.28	1736.85	94.0812	1634.05	100.66	1.59

neither declared nor measured, simulations of the PAN shuffler response to each were performed at five different densities (2.4 g/cm³, 3.0 g/cm³, 3.6 g/cm³, 4.2 g/cm³, and 4.8 g/cm³). Table 3 summarizes the comparison of MCNP simulated and measured delayed neutron count rates for the LLNL developed PuO₂ WRMs at an assumed density of 2.4 g/cm³.

At an assumed PuO₂ density of 2.4 g/cm³, the maximum and minimum in the ratios of the simulated to measured count rates are $1.264 \pm 28.26\%$ (PuOKC0040) and $0.922 \pm 5.97\%$ (PuOKC0810), respectively, and the average of the ratios is $1.037 \pm 9.35\%$. With an increase in PuO₂ density to 4.8 g/cm³, the maximum and minimum in the ratios decrease to $1.263 \pm 28.23\%$ (PuOKC0040) and $0.910 \pm 5.96\%$ (PuOKC0810), respectively, and the average of the ratios decreases approximately 0.9% to $1.028 \pm 9.37\%$. While the appropriateness of Monte Carlo simulations as a tool in the PAN shuffler calibration process was validated as part of the LLNL HEU oxide calibration [1], considering the fact that the PuO₂ WRMs were developed from the well characterized LLNL inventory of PuO₂-bearing containers, the above results can only serve to add further credence to the conclusion.

Monte Carlo Simulations of PAN Shuffler Response Sensitivity to Variations in PuO₂ Density and Container Diameter

A significant part of the effort to determine the sensitivity of the PAN shuffler response to variations in PuO₂ density and container diameter was accomplished with the MCNP simulations of the PAN shuffler response to the PuO₂ WRMs. To complete the effort, an extensive series of additional MCNP simulations were performed. Response sensitivities to variations in PuO₂ density and container diameter were evaluated for the

Table 3. Comparison of MCNP simulated and measured delayed neutron count rate results for LLNL PuO₂ WRMs at an assumed density of 2.4 g/cm³.

LLNL Nomenclature	Simulated count rate		Measured count rate (counts/s)	Simulated to measured ratio
	²³⁹ Pu (counts/s)	^{238, 239, 240, 241} Pu and ²⁴¹ Am ^a (counts/s)		
PuOKC0040	36.87 ± 0.47	37.64 ± 0.47	29.78 ± 8.41	1.264 ± 28.26%
PuOKC0080	59.78 ± 0.63	61.20 ± 0.63	60.72 ± 5.98	1.008 ± 9.90%
PuOSQ0110	80.10 ± 0.72	81.64 ± 0.72	65.59 ± 3.97	1.245 ± 6.12%
PuOKC0160	103.27 ± 0.85	106.45 ± 0.85	100.28 ± 8.16	1.062 ± 8.17%
PuOKC0210	132.90 ± 0.98	136.75 ± 0.98	139.03 ± 10.33	0.984 ± 7.47%
PuOKC0240	150.71 ± 1.05	155.31 ± 1.05	157.00 ± 17.26	0.989 ± 11.01%
PuOSQ0340	208.18 ± 1.27	214.46 ± 1.27	211.40 ± 5.39	1.014 ± 2.62%
PuOKC0420	244.94 ± 1.39	252.86 ± 1.39	249.79 ± 27.22	1.020 ± 11.00%
PuOSQ0610	358.96 ± 1.79	368.03 ± 1.79	387.99 ± 17.12	0.949 ± 4.44%
PuOSQ0800	461.82 ± 2.09	478.95 ± 2.09	483.96 ± 21.63	0.990 ± 4.49%
PuOKC0810	459.06 ± 2.05	475.23 ± 2.06	515.59 ± 30.71	0.922 ± 5.97%
PuOKC1010	566.06 ± 2.35	586.82 ± 2.35	606.29 ± 20.55	0.968 ± 3.41%
PuOSQ1090	618.66 ± 2.52	639.44 ± 2.52	612.72 ± 41.37	1.037 ± 6.72%
PuOKC1250 ^b	695.36 ± 2.75	721.39 ± 2.75	739.19 ± 34.04	0.976 ± 4.62%
PuOSQ1410	786.98 ± 2.94	813.41 ± 2.94	756.29 ± 9.17	1.076 ± 1.27%
PuOSQ1640	910.77 ± 3.26	941.29 ± 3.26	915.08 ± 28.59	1.029 ± 3.14%
			Average	1.037 ± 9.35%

a. ^{239, 240, 241}Pu isotopes account for 99.9% of the total count rate.

b. Results are for a density of 3.0 g/cm³. Material quantity exceeds the container capacity at a density of 2.4 g/cm³.

five different densities (2.4 g/cm³, 3.0 g/cm³, 3.6 g/cm³, 4.2 g/cm³, and 4.8 g/cm³) and two different primary containers (Kaufman can and quart can) using PuO₂ of an elemental and isotopic composition equal to the average of the PuOKC (93.9246 wt% ²³⁹Pu) and PuOSQ (94.2295 wt% ²³⁹Pu) WRMs and mass range that varied in irregular increments from a minimum of 10 g to the nominal maximum for the primary container and density under evaluation.

As was the case with the models of the PuO₂ WRMs, the sensitivity models used in the MCNP simulations were true to the packaging of each, including the primary containers, packing materials (polyvinylchloride bag-out bag and polyethylene poultry bag), and secondary (over-pack) containers required to satisfy LLNL Plutonium Facility containment requirements.

PuO₂ Calibration Algorithms and Error Modeling

The MCNP simulations give separate information for delayed neutrons from each isotope and for the detector efficiency. In parallel to the situation for U₃O₈ irradiation [1], there are effects of attenuation, self-shielding, and multiplication in the PuO₂ containers as well. Hence the linear density of ²³⁹Pu in the container, as

described by Eq. 4 in [1], is used as a key variable. Figure 1 shows the ^{239}Pu delayed neutron production rate per unit mass versus ^{239}Pu linear density. The curves versus linear density are close to a common curve and are fitted with a single model, which sums three declining exponentials and a constant. The simulation results and model agree within a standard deviation of 0.4%. The results for ^{240}Pu and ^{241}Am increase with ^{239}Pu linear density, similar to the case for ^{238}U in U_3O_8 . The same form of model is used. The variation among the ^{240}Pu curves versus ^{239}Pu linear density has a standard deviation from a common curve of 1.5%, and is up to $\pm 3\%$ in some cases, varying with PuO_2 density. The contribution of ^{240}Pu to the total Pu delayed neutron counts is only 2% to 4%, hence the common model is used for the ^{240}Pu curves. Detector efficiency rises with ^{239}Pu linear density, similar to the case for U_3O_8 but with about three times the slope. The variation of the curves from a common model curve has a standard deviation of 0.44%.

For initial application, only the lower PuO_2 or mixed U-Pu oxide densities, primarily 2.4 g/cm^3 and in some cases ranging up to 3.3 g/cm^3 , are considered. Hence the corresponding Pu model is based on the simulation results only at the densities of 2.4 and 3.0 g/cm^3 . Then the model variations have standard deviations of 0.35%, 0.8%, and 0.22% for ^{239}Pu , ^{240}Pu , and efficiency, respectively. The small contributions from ^{241}Pu and ^{241}Am are accounted for as curves proportional to the ^{241}Pu and ^{240}Pu curves, respectively. The overall Pu model uncertainty is 0.41% for fitting and 3.7% for model calibration (see Table 3), for a total of 3.7%.

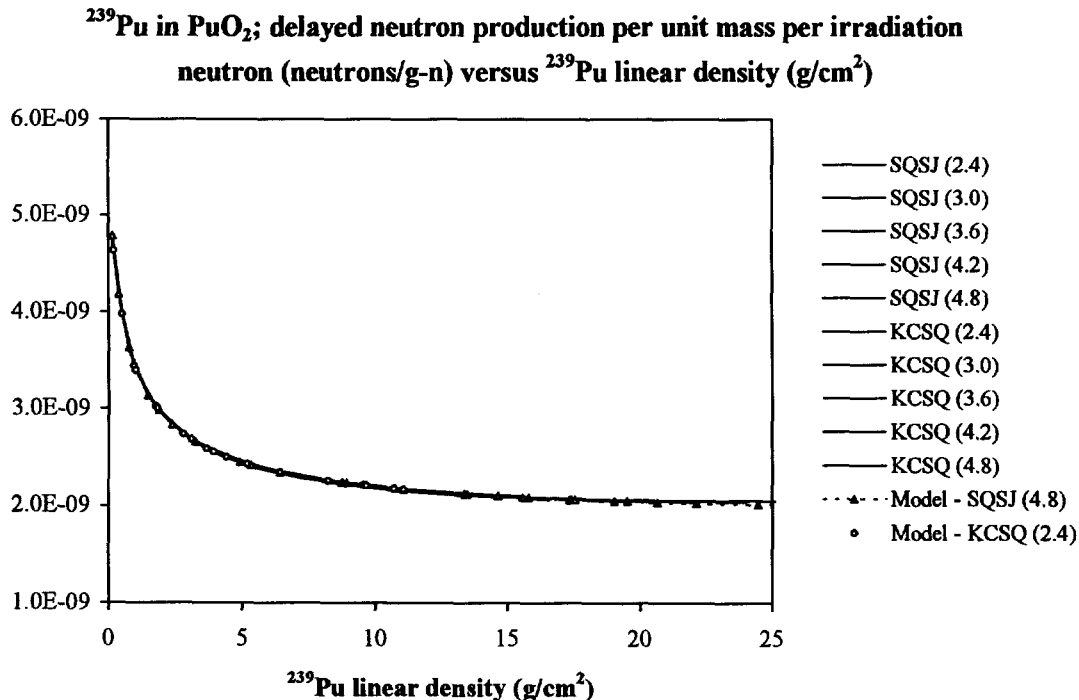


Figure 1. Delayed neutron production from only the ^{239}Pu in a sample of PuO_2 , per unit mass of ^{239}Pu , versus ^{239}Pu linear density, for reference Pu isotopic composition, for selected container conditions. In the chart legend, the first text item identifies the primary container (SQSJ means a quart container and KCSQ means a Kaufman can) and the second text item identifies the PuO_2 density in g/cm^3 .

Mixed U-Pu Oxide Calibration Algorithms and Error Modeling

A further series of MCNP simulations was performed for mixed U-Pu oxides at a density of 2.4 g/cm^3 , in the Kaufman can and quart can, in steps of 10% from a 10% PuO_2 – 90% U_3O_8 mixture to a 90% PuO_2 – 10% U_3O_8 mixture, for a full range of mixed U-Pu oxide masses which can be contained within the respective cans.

Here the delayed-neutron production curves for the isotopes appear similar to their curves for separate PuO_2 or U_3O_8 . Effects of attenuation, self-shielding, and multiplication are apparent. The question arises as to how the added isotopes add to or subtract from these effects. The answers found are fairly simple from a modeling perspective. The ^{235}U self-shielding is influenced mainly by the ^{235}U , with some contribution from ^{239}Pu . Similarly the ^{239}Pu self-shielding is influenced mainly by the ^{239}Pu , with some contribution from ^{235}U . For the fissionable isotopes ^{238}U and ^{240}Pu , the increase in delayed neutron production is influenced more by the ^{239}Pu present and to a smaller extent by the ^{235}U . The same proportionality between ^{235}U influence and ^{239}Pu influence is found for both the ^{238}U and the ^{240}Pu . The efficiency curve is a straight line and the parameters can be interpolated linearly between the U_3O_8 result and the PuO_2 result.

Figure 2 shows the ^{235}U delayed neutron production rate per unit mass of ^{235}U , in the quart container, versus a combined linear density $\text{CD}_{\text{L}_{235\text{U}}}$, in g/cm^2 , of

$$\text{CD}_{\text{L}_{235\text{U}}} = 0.6 \times (\text{D}_{\text{L}_{239\text{Pu}}}) + 1.0 \times (\text{D}_{\text{L}_{235\text{U}}}) \quad (1)$$

where $\text{D}_{\text{L}_{239\text{Pu}}}$ and $\text{D}_{\text{L}_{235\text{U}}}$ are the linear densities of ^{239}Pu and ^{235}U , in g/cm^2 , respectively, as described by Eq. 4 in [1].

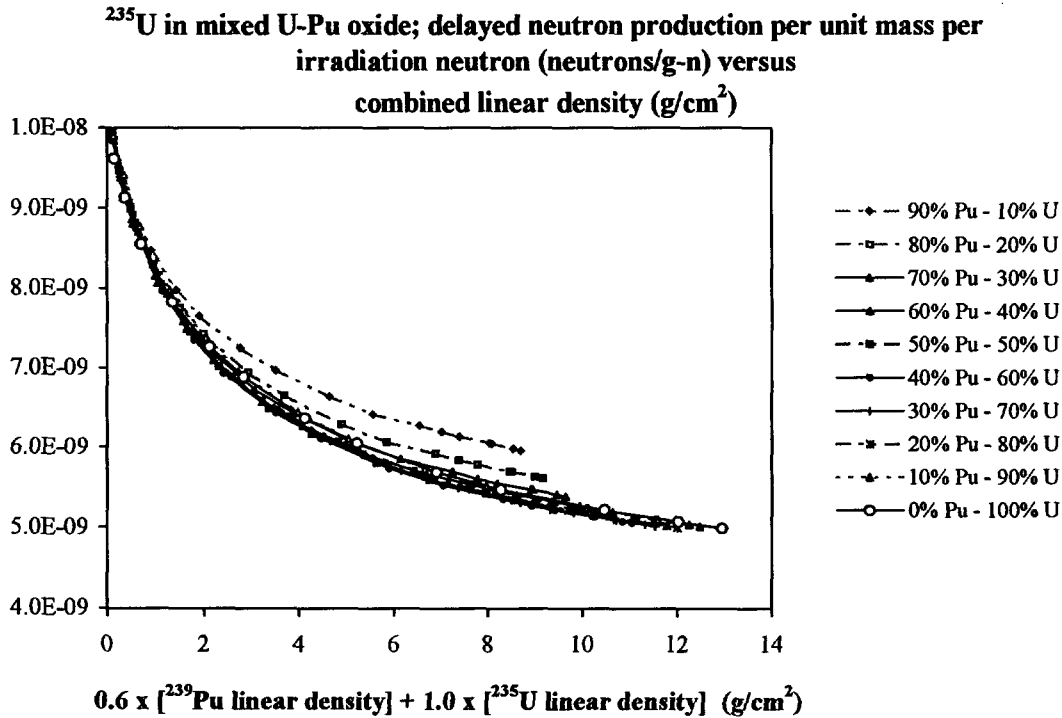


Figure 2. Delayed neutron production from only the ^{235}U in a sample of mixed U-Pu oxide, per unit mass of ^{235}U , versus a combined linear density, for a series of PuO_2 -to- U_3O_8 proportions in the quart container at a mixed U-Pu oxide density of 2.4 g/cm^3 , and for the summary model of ^{235}U delayed neutrons for 100% U_3O_8 at the density of 2.4 g/cm^3 and for the same container.

The curve for 100% U_3O_8 has been multiplied by a factor of 1.112 to account for the different irradiation time cycle used in the mixed U-Pu oxide irradiation and modeling. This factor is slightly different from the measured factor of 1.121 described above and is found by matching up the 100% U_3O_8 curve with the series of mixed U-Pu oxide curves versus mixed U-Pu oxide or U_3O_8 mass, for the limited range of containers and densities used in the mixed U-Pu oxide model application. Most of the mixed U-Pu oxide curves line up closely with the curve for the case of 100% U_3O_8 . There is a variation for mixtures with 80% and 90% Pu.

For mixtures greater than 70% Pu, a coefficient of $D_{L_{239Pu}}$ varying with %Pu, rather than the constant 0.6, is used in the above equation. This adjustment will line up the curves for ^{235}U in 80% Pu and 90 % Pu mixed U-Pu oxide with the curve for 100% U_3O_8 . The curves for the Kaufman can show the same results.

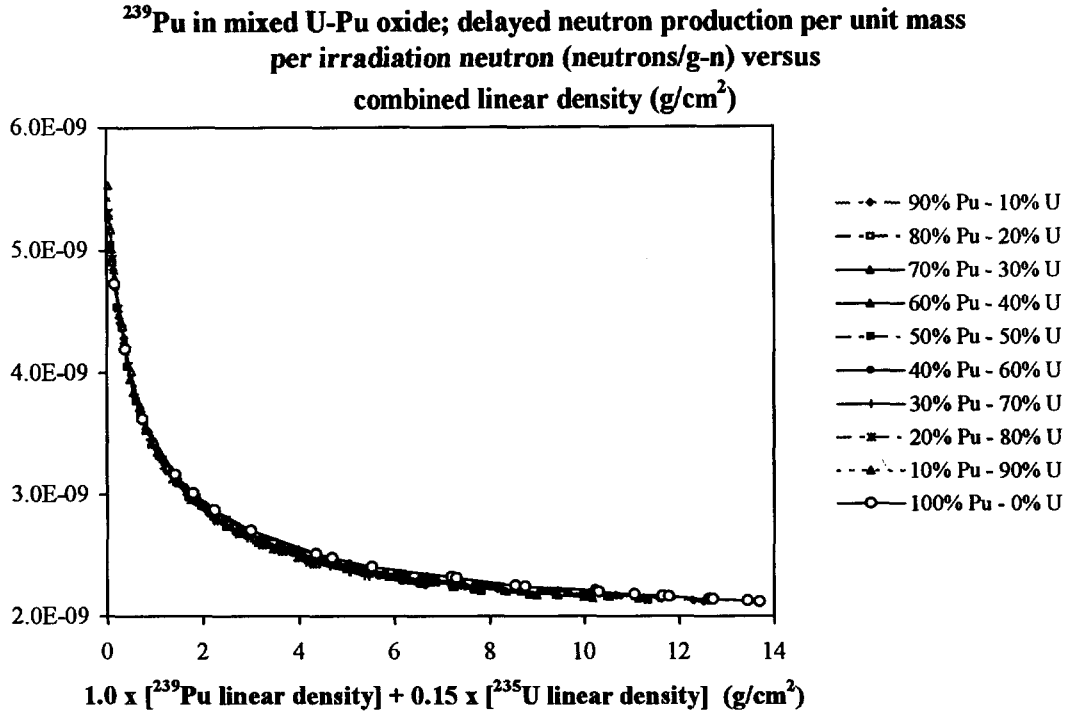


Figure 3. Delayed neutron production from only the ^{239}Pu in a sample of mixed U-Pu oxide, per unit mass of ^{239}Pu , versus a combined linear density, for a series of PuO_2 -to- U_3O_8 proportions in the quart container at a mixed U-Pu oxide density of $2.4 g/cm^3$, and for the MCNP simulations of ^{239}Pu delayed neutrons for 100% PuO_2 at the density of $2.4 g/cm^3$ and for the same container.

Figure 3 shows the ^{239}Pu fission production rate per unit mass of ^{239}Pu , in the quart container, versus a combined linear density $CD_{L_{239Pu}}$, in g/cm^2 , of

$$CD_{L_{239Pu}} = 1.0 \times (D_{L_{239Pu}}) + 0.15 \times (D_{L_{235U}}) \quad (2)$$

and all other terms are as previously defined. The curves line up closely with the MCNP simulation curve shown (and the model curve) for the case of 100% PuO_2 . The curves for the Kaufman can show the same results.

Figure 4 shows the ^{240}Pu fission production rate per unit mass of ^{240}Pu , in the quart container, versus a combined linear density $CD_{L_{240Pu}}$, in g/cm^2 , of

$$CD_{L_{240Pu}} = 1.0 \times (D_{L_{239Pu}}) + 0.55 \times (D_{L_{235U}}) \quad (3)$$

and all other terms are as previously defined. The curves line up closely with the curve for the case of 100% PuO_2 . There are minor variations which are not significant to the total result because the ^{240}Pu contributes only 2% to 4% of the total Pu counts. Again, the curves for the Kaufman can show the same results.

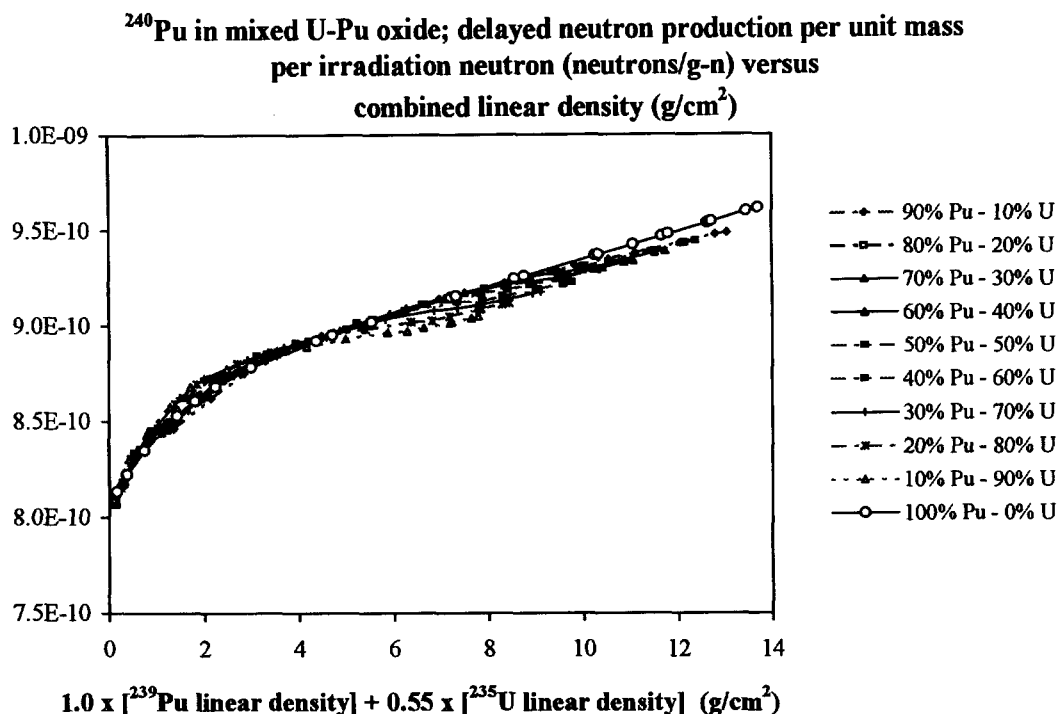


Figure 4. Delayed neutron production from only the ^{240}Pu in a sample of mixed U-Pu oxide, per unit mass of ^{240}Pu , versus a combined linear density, for a series of PuO_2 -to- U_3O_8 proportions in the quart container at a mixed U-Pu oxide density of $2.4 \text{ g}/\text{cm}^3$, and for the summary model of ^{240}Pu delayed neutrons for 100% PuO_2 at the density of $2.4 \text{ g}/\text{cm}^3$ and for the same container.

The algorithms for the calibration curves follow this curve-matching procedure.

Selected Unknown Item Measurement Results

Table 4 summarizes the measurement results for a selected set of LLNL unknown mixed U-Pu oxide items chosen to demonstrate the applicability of the LLNL PAN shuffler mixed U-Pu oxide calibration algorithms. These items cover the evaluated range in container diameter (8.57 cm to 9.88 cm), an extensive range in measured Pu mass (0.95 g to 1528.89 g), an extensive range in declared U mass (12 g to 1479 g), and an extensive range in declared ^{235}U enrichment (40.00 wt% ^{235}U to 96.46 wt% ^{235}U). Because the mixed U-Pu oxide density of each was neither declared nor measured, all items were initially assumed to have a nominal mixed U-Pu oxide of $2.4 \text{ g}/\text{cm}^3$. However, for those items whose measured U-Pu oxide mass was calculated to exceed the capacity of its container, the U-Pu oxide density was increased in increments of $0.1 \text{ g}/\text{cm}^3$ to the extent necessary for the container to accommodate the measured U-Pu oxide mass.

Because of the very nature of the items themselves (i.e., unknowns with measured values for Pu mass and experimenter declared values for U mass), the spread exhibited in the individual differences between the declared and measured U mass and the total U mass difference is not unexpected. While the measured U mass results reflect the accountability values for these items and are therefore not subject to an inventory difference analysis, the total U mass difference of 26.54 g is no more than 0.44% of the total measured U mass and well within the standard deviation ($\pm 92.18 \text{ g}$) and the 95% confidence limit ($\pm 182.51 \text{ g}$) in the total measured U mass.

Table 4. Summary of measurement results for selected LLNL U-Pu oxide items.

Item Identification	Container diameter (cm)	U-Pu oxide density (g/cm ³)	Measured	Declared		Measured U mass (g)	U mass difference (g)
			Pu mass ¹ (g)	U mass (g)	²³⁵ U wt% (%)		
MRF003956	8.57	2.4	380.25	16.12	93.18	10.96 ± 10.07 ²	-5.16
MRF003952	8.57	2.4	193.22	33.86	96.46	20.90 ± 11.10	-12.96
MRF003858	8.57	2.4	795.71	317.00	93.83	274.79 ± 20.79	-42.21
MRF003877	8.57	2.4	248.96	348.00	94.02	348.15 ± 11.02	0.15
MRF003861	8.57	2.4	605.63	531.00	95.00	425.76 ± 19.18	-105.24
MRF003870	8.57	2.4	484.22	675.00	94.00	655.44 ± 20.38	-19.56
MRF003872	8.57	2.8	298.71	1087.20	94.70	999.09 ± 23.47	-88.11
MRF003887	8.57	3.3	52.82	1479.00	93.13	1461.03 ± 30.25	-17.97
MRF003959	9.88	2.4	363.57	12.00	93.00	63.77 ± 20.62	51.77
MRF003953	9.88	2.4	1528.89	15.65	93.16	20.40 ± 29.74	4.75
MRF003960	9.88	2.4	258.95	36.00	40.00	140.30 ± 46.82	104.30
MRF003961	9.88	2.4	157.99	42.00	69.48	73.51 ± 22.32	31.51
MRF003967	9.88	2.4	11.63	60.49	94.01	46.96 ± 5.91	-13.53
MRF003951	9.88	2.4	203.75	90.80	93.16	90.85 ± 8.14	0.05
MRF003934	9.88	2.4	343.91	176.00	80.56	303.77 ± 29.97	127.77
MRF003928	9.88	2.4	0.95	452.79	93.16	476.48 ± 9.61	23.69
MRF003869	9.88	2.4	493.60	576.91	93.16	564.2 ± 19.42	-12.71
Total						5976.36 ± 92.18	26.54

1. Decayed to the date of the PAN shuffler measurement.
2. Average measured U-Pu oxide count rate yields a negative algorithm solution for measured U mass. Results are based on the average measured U-Pu oxide count rate plus $1.96 \times \sigma_{\text{Meas}}$ (95% confidence limit).

References

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